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**Polyester Resin Composition**

**Your Ref: 103003 - 11**

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(54) [Title of the Invention]

**Polyester Resin Composition**

(57) [Summary]

[Object] To provide a readily calenderable polyester resin composition that is of superior roll release properties, long run capacity, and thermal stability.

[Means of Achievement] A readily calenderable polyester resin composition is obtained by means of compounding one or more aliphatic phosphoric acid ester compounds or by means of

further compounding one or more phenolic antioxidants and/or organic phosphorous acid ester compounds with a polyester resin.

**[Claims]**

**[Claim 1]** A polyester resin composition for use in calender molding, characterized in that at least one substance selected from aliphatic phosphoric acid fatty acid ester compounds is compounded with a polyester resin.

**[Claim 2]** A polyester resin composition for use in calender molding as described in Claim 1, wherein at least one substance selected from phenolic antioxidants and organic phosphorous acid ester compounds is further used in combination with the composition.

**[Claim 3]** A polyester resin composition for use in calender molding as described in Claim 1, wherein at least one substance selected from organic phosphoric acid ester compounds is used in combination with the resin composition described in Claim 1.

**[Claim 4]** A polyester resin composition for use in calender molding as described in Claim 3, wherein at least one substance selected from phenolic antioxidants and organic phosphorous acid ester compounds is further used in combination with the resin composition described in Claim 3.

**[Detailed Description of the Invention]**

**[0001]**

**[Technological Field of the Invention]** The present invention relates to the calendering of polyester resins, and, in further detail, provides polyester resins that are of superior roll release properties, long run capacity, and thermal stability, and that are useful for various types of sheets and films, such as shrink labels for food products, cosmetic products and beverages, fragrance-retaining heat-seal films, and oil resistant multilayer sheets.

**[0002]**

**[Prior Art]** Because of the ease of processing, extrusion molding has been used primarily as a method for manufacturing sheets and films of polyester resins. In comparison with extrusion molding, however, calender molding is less precise in terms of film thickness, width, direction, flow direction, and other attributes because the process depends on the friction of the die lips. Nor is calendering suited to large volume production, and a need therefore exists for developing a polyester resin that can be calender molded.

[0003] When thermoplasticized, polyester resins adhere strongly to the rolls, and various lubricants have therefore been studied. For example, studies have been conducted concerning various types of lubricants, including hydrocarbon lubricants such as polyethylene wax and paraffin wax, high fatty acid lubricants, metallic soaps based on higher fatty acids, fatty acid amide lubricants, and ester lubricants. However, it has not been possible to obtain sufficient roll release properties and long run time.

[0004]

**[Problems to Be Solved by the Invention]** This invention addresses the problems of the prior art and provides a polyester resin that has superior roll release properties, long run capacity, and thermal stability and that is optimum for calender processing.

[0005]

**[Means Used to Solve the Above-Mentioned Problems]** The means for solving the aforementioned problems is as follows. A polyester resin that possesses superior roll release properties, long run capacity, and thermal stability and that is optimum for calendering can be obtained by means of compounding at least one aliphatic phosphoric acid ester compound with a polyester resin, and, further, by means of the combined use of organic phosphoric acid ester compounds, phenolic antioxidants, and/or organic phosphorous acid ester compounds that are different from the above.

[0006] As used herein, "polyester resin" refers to polyethylene terephthalate, polybutylene terephthalate, a copolymer of terephthalic acid and a mixed alcohol of ethylene glycol and 1,4-dihydroxymethyl cyclohexane, poly-1,4-cyclohexane dimethylene terephthalate, or polybutylene naphthalate, which may be used singly or as mixtures of two or more polyester resins or as mixtures with other thermoplastic resins, such as polycarbonates, ABS, polypropylenes, polyethylene, nylon, and rubber, with the polyester resin being the principal component.

[0007] The term "aliphatic phosphoric acid ester compound" used in relation to this invention refers to a mono-, di-, or triester compound of phosphoric acid with a saturated or unsaturated higher aliphatic alcohol having 6 to 30 carbon atoms, or a metal salt thereof. Typical examples are presented here, but the ester compound is not limited to them. The aliphatic phosphoric acid ester compound that can be used may be triisodecyl phosphate, tridodecyl phosphate, trihexa-

decyl phosphate, trioctadecyl phosphate, dihexyl octadecyl phosphate, decyl dodecyl tridecyl phosphate, dinonyl 1-methylpentyl phosphate, bis(2-methylhexyldecyl) pentadecyl phosphate, eicosyl bis(12-tridecyl) phosphate, tris(cis-9-octadecyl) phosphate, cis-9-octadecyl dioctadecyl phosphate, ditetradecyl phosphate, diheptyl phosphate, dioctadecyl phosphate, didodecyl phosphate, nonyl tetradecyl phosphate, cis-9-octa decyl octadecyl phosphate, hexadecyl decyl phosphate, dieicosyl phosphate, bis(cis-9-octadecyl) phosphate, dodecyl phosphate, octadecyl phosphate, eicosyl phosphate, or octyl phosphate, or a lithium, sodium, potassium, magnesium, calcium, barium, zinc, or aluminum metal salt thereof. The quantity of the aliphatic phosphoric acid ester compound added should be 0.05 to 5.0 parts by weight, and, preferably, 0.1 to 2.0 parts by weight per 100 parts by weight of polyester resin. When the quantity added is less than 0.05 parts by weight, roll lubricity is insufficient. When the quantity is greater than 5.0 parts by weight, rising may occur on the surface, which is an undesirable occurrence.

[0008] Next, phenolic antioxidants that can be used in this invention will be described.

However, the antioxidants are not limited to this list. Additional examples include 2,6-di-tert-butyl phenol, 2,6-di-tert-butyl-4-methyl phenol, 2,6-di-tert-butyl-4-ethyl phenol, 2-tert-butyl-4,6-dimethyl phenol, 2,4,6-tri-tert-butylphenol, 2-tert-butyl-4-methoxyphenol, 3-methyl-4-isopropyl phenol, 2,6-di-tert-butyl-4-hydroxymethyl phenol, 2,2-bis(4-hydroxyphenyl) propane, bis(5-tert-butyl-4-hydroxy-2-methylphenyl) sulfide, 2,5-di-tert-amyl hydroquinone, 2,5-di-tert-butyl hydroquinone, 1,1-bis(3-tert-butyl-4-hydroxy-5-methylphenyl) butane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl) methane, 2,6-bis(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methyl phenol, bis(3-tert-butyl-4-hydroxy-5-methylbenzyl) sulfide, bis(3-tert-butyl-5-ethyl-2-hydroxyphenyl) methane, bis(3,5-di-tert-butyl-4-hydroxyphenyl) methane, bis(3-tert-butyl-2-hydroxy-5-methylphenyl) sulfide, 1,1-bis(4-hydroxyphenyl) cyclohexane, ethylene bis[3,3-bis(3-tert-butyl-4-hydroxyphenyl)butyrate], bis[2-(2-hydroxy-3-tert-butyl-5-methylbenzyl)-4-methyl-6-tert-butylphenyl] terephthalate, 1,1-bis(2-hydroxy-3,5-dimethylphenyl)-2-methyl propane, 4-methoxyphenol, cyclohexyl phenol, p-phenyl phenol, catechol, hydroquinone, 4-tert-butyl pyrocatechol, ethyl gallate, propyl gallate, octyl gallate, lauryl gallate, cetyl gallate,  $\beta$ -naphthol, 2,4,5-trihydroxybutyrophenone, tris(3,5-di-tert-butyl-4-hydroxyphenyl) isocyanate, 1,3,5-trimethyl-2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl) benzene, 1,6-bis[2-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxy] hexane, tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionyloxymethyl] methane, bis(3-cyclohexyl-2-hydroxy-5-methylphenyl) methane, bis[3-

(3,5-di-tert-butyl-4-hydroxyphenyl)propionyloxyethyl] sulfide, n-octadecyl-3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate, bis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionylamino] hexane, 2,6-bis(3-tert-butyl-2-hydroxy-5-methylphenyl)-4-methylphenol, bis[S-(4-tert-butyl-3-hydroxy-2,6-di-methylbenzyl)] thioterephthalate, tris[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyl-oxyethyl] isocyanurate, and 1,1,3-tris(3-tert-butyl-4-hydroxy-6-methylphenyl) butane. The quantity in which these phenolic antioxidants are added should be 0.01 to 1.0 parts by weight, and, preferably, 0.02 to 0.8 parts by weight, per 100 parts by weight of polyester resin. When the quantity added is less than 0.01 parts by weight, there is poor thermal stability during molding processing, for which reason coloration of the resin and adhesion to the roll due to thermal deterioration occur. Addition of more than 1.0 part by weight is not economical because the effect of the invention is saturated.

[0009] Next, examples of the organic phosphorous acid ester compounds used in this invention will be described. However, other compounds may also be used. Additional examples include triphenyl phosphite, tris(methylphenyl) phosphite, triisooctyl phosphite, tridecyl phosphite, tris(2-ethylhexyl) phosphite, tris(nonylphenyl) phosphite, tris(octylphenyl) phosphite, tris[decyl poly(oxyethylene) phosphite, tris(cyclohexylphenyl) phosphite, tricyclohexyl phosphite, tri(decyl) thiophosphite, triisodecyl thiophosphite, phenyl bis(2-ethylhexyl) phosphite, phenyl diisodecyl phosphite, tetradecyl poly(oxyethylene) bis(ethylphenyl)phosphite, phenyl dicyclohexyl phosphite, phenyl diisooctyl phosphite, phenyl di(tridecyl) phosphite, diphenyl cyclohexyl phosphite, diphenyl isooctyl phosphite, diphenyl 2-ethylhexyl phosphite, diphenyl isodecyl phosphite, diphenyl cyclohexylphenyl phosphite, diphenyl (tridecyl) thiophosphite, nonylphenyl ditridecyl phosphite, phenyl p-tert-butylphenyl dodecyl phosphite, diisopropyl phosphite, bis[octadecyl poly(oxyethylene)] phosphite, octyl poly(oxypropylene) tridecyl poly(oxypropylene) phosphite, monoisopropyl phosphite, diisodecyl phosphite, diisooctyl phosphite, monoisooctyl phosphite, didodecyl phosphite, monododecyl phosphite, dicyclohexyl phosphite, monocyclohexyl phosphite, monododecyl poly(oxyethylene) phosphite, bis(cyclohexylphenyl) phosphite, monocyclohexyl phenyl phosphite, bis(p-tert-butylphenyl) phosphite, tetratridecyl 4,4'-isopropylidene diphenyl diphosphite, tetratridecyl 4,4'-butylidene bis(2-tert-butyl-5-methylphenyl) diphosphite, tetraisooctyl 4,4'-thiobis(2-tert-butyl-5-methylphenyl) diphosphite, tetrakis(nonylphenyl) poly-(propyleneoxy) isopropyl diphosphite, tetratridecyl propyleneoxypropyl diphosphite, tetratridecyl 4,4'-isopropylidene dicyclohexyl diphosphite,

pentakis(nonylphenyl) bis[poly(propyleneoxy)isopropyl] triphosphite, heptakis(nonylphenyl) tetrakis(propyleneoxy)isopropyl] pentaphosphite, heptakis(nonylphenyl) tetrakis(4,4'-isopropylidene diphenyl) pentaphosphite, decakis(nonylphenyl) heptakis(propylene-oxyisopropyl) octaphosphite, decaphenyl heptakis(propyleneoxyisopropyl) octaphosphite, bis(carbobutoxyethyl) 2,2-dimethylene-trimethylene dithiophosphite, bis(isooctoxycarbomethyl 2,2-dimethylene trimethylene dithiophosphite, tetradodecyl ethylene dithiophosphite, tetradodecyl hexamethylene dithiophosphite, tetradodecyl 2,2'-oxydiethylene dithiophosphite, pentadodecyl di(hexamethylene) trithiophosphite, diphenyl phosphite, 4,4'-isopropylidene-dicyclohexyl phosphite, 4,4'-isopropylidene-diphenyl alkyl (C12-C15) phosphite, 2-tert-butyl-4-[1-(3-tert-butyl-4-hydroxyphenyl-isopropyl)phenyl di(p-nonylphenyl) phosphite, ditridecyl 4,4'-butylidene bis(3-methyl-6-tert-butylphenyl) phosphite, dioctadecyl 2,2-dimethylene trimethylene diphosphite, tris-(cyclohexylphenyl) phosphite, hexatridecyl 4,4',4''-1,1,3-butane tolyl-tris(2-tert-butyl-5-methylphenyl) triphosphite, tridodecyl thiophosphite, decaphenyl heptakis(propyleneoxyisopropyl) octaphosphite, dibutyl pentakis(2,2-dimethylene trimethylene) diphosphite, dioctyl pentakis(2,2-dimethylene trimethylene) diphosphite, and didecyl 2,2-dimethylene trimethylene diphosphite, as well as lithium, sodium, potassium, magnesium, calcium, barium, zinc, and aluminum metal salts thereof. The quantity in which these organic phosphorous acid ester compounds are added should be 0.01 to 3.0 parts by weight, and, preferably, 0.02 to 2.0 parts by weight, per 100 parts by weight of polyester resin. When the quantity added is less than 0.01 parts by weight, there is poor thermal stability during molding, for which reason coloration of the resin and adhesion to the roll due to thermal deterioration occur. Addition of more than 3.0 parts by weight is not economical because the effect of the invention is saturated. These phenolic antioxidants and organic phosphorous acid ester compounds may be used alone. However, when two or more are used in combination, a more pronounced effect can be obtained.

**[0010]** Even when the aliphatic phosphoric acid ester compounds are used individually in this invention, their efficacy is clearly manifested. However, when organic phosphorous acid ester compounds different from these compounds are used in conjunction with these compounds, a much more pronounced effect is obtained. These organic phosphoric acid ester compounds are organic phosphoric acid ester compounds for which alcohols containing ether groups are used as raw materials and which are esterified using at least one alcohol as a component. It may contain an alcohol or phenol compound devoid of ether groups as another alcohol component. It may



also be a free substance in which a hydroxy group bonded to the phosphoric acid does not react with the alcohol. Representative examples are listed below. However, they are not limited to this list. The organic phosphoric acid ester compounds that can be used include tris[methyl di(oxyethylene) phosphate, tris[butyl di(oxyethylene)] phosphate, tris(2-methoxypropyl) phosphate, tris(hexoxyethyl) phosphate, tris[ethyl di(oxypropylene)] phosphate, tris-[butyl poly(oxypropylene)] phosphate, tris[octyl poly(oxypropylene)] phosphate, tris-[phenyl poly(oxypropylene)] phosphate, tris[dodecyl poly(oxyethylene)] phosphate, tris-[2-(2-hydroxyethoxy)-ethyl] phosphate, bis[ethyl di(oxyethylene)] 2-(2-hydroxypropoxy) propyl phosphate, bis[methyl di(oxyethylene)] butyl poly(oxypropylene) phosphate, didodecyl ethyl di(oxypropylene) phosphate, tridecyl 2-methoxypropyl 2-hydroxyethyl di(oxyethylene) phosphate, bis-[methyl di(oxyethylene)] tolyl phosphate, octyl ethyl di(oxyethylene) nonylphenyl phosphate, tetrakis(2-methoxyethyl) 2,2'-oxydiethyl diphosphate, tetrakis[ethyl di(oxy-ethylene)] 2,2'-oxybis(1-methylethyl)] 2,2'-oxybis(1-methylethyl)) diphosphate, tetrakis[acetyl di(oxyethylene)] 5,5'-oxybis(3-oxapentamethylene) diphosphate, bis-[methyl tri(oxyethylene)] bis(2-methoxyethyl) O,O'-propylene di(oxypropylene) diphosphate, methyl di(oxyethylene) ethyl tri(oxypropylene) 2,2'-dimethylene trimethylene) diphosphate, tri(octadecyl) 2-octoxyethyl trimethylene diphosphate, bis[methyl di(oxypropylene) bis[ethyl tri(oxyethylene)] 4,4'-methylene diphenyl phosphate, bis[tetradecyl poly(oxyethylene)] phosphate, bis[methyl di(oxyethylene)]-phosphate, bis(phenoxyethyl) phosphate, bis[2-hydroxypropyl poly(oxypropylene)] phosphate, hexadecyl 2-hydroxyethoxyethyl phosphate, cyclohexyl phenylpropyl di(oxyethylene)] ethylene poly(oxyethylene) diphosphate, bis[hexyl poly(oxyethylene)] ethylene poly(oxyethylene) diphosphate, propyl di(oxypropylene) 2-hydroxypropyl di(oxypropylene) phosphate, bis[hydroxypropyl poly(oxypropylene) phosphate, dodecyl 2-hydroxyethyl poly(oxyethylene) 4,4'-thiodiphenyl diphosphate, methyl di(oxyethylene) phosphate, 2-hydroxypropyl di(oxypropylene) phosphate, 2-hydroxypropyl poly(oxypropylene) phosphate, and octadecyl poly(oxyethylene) phosphate, as well as lithium, sodium, potassium, magnesium, calcium, barium, zinc, and aluminum metal salt compounds thereof. The quantity in which these organic phosphoric acid esters are added should be 0.05 to 5.0 parts by weight, and, preferably, 0.1 to 3.0 parts by weight, per 100 parts by weight of polyester resin. When the quantity is less than 0.05 parts by weight, a sufficient effect of combined use is not obtained when these esters

are used in combination with aliphatic phosphoric acid ester compounds. When the compound exceeds 5.0 parts by weight, the function/cost balance is adversely affected.

[0011] Polyhydric alcohols can be added to the polyester resin of this invention as needed.

Polyhydric alcohols that can be used in this invention include, for example, pentaerythritol, dipentaerythritol, trimethylolpropane, ditrimethylol propane, and dipentaerythritol adipic acid ester.

[0012] Other components may be added as needed in an appropriate manner to the polyester resin of this invention. For example, fillers, ultraviolet absorbers, photostabilizers, pigments, antistatic agents, antimicrobial agents, epoxy compounds, crosslinking agents, and sulfur antioxidants can be cited.

[0013] We shall now describe this invention in greater detail on the basis of working examples.

[0014] The various components of Working Examples 1 to 17 and Comparatives Examples 1 to 6 as shown in Table 1 were mixed, the sheet thickness was set to 0.3 mm in six-inch chilled rolls heated to 180°C and 190°C, and adhesion time to the rolls was determined. The results are shown in Table 1.

[0015]

[Table 1]

		Working Examples											
No.		1	2	3	4	5	6	7	8	9	10	11	12
A		100	100	100	100	100	100	100	100	100	100	100	100
B		0.5			0.5	0.3		0.2	0.2	0.5	0.5		
C			0.5			0.2	0.3	0.3	0.3			0.5	0.5
D				0.5			0.2						
E					0.1			0.1	0.1	0.1			0.1
F						0.1			0.1				
G						0.1				0.1			
H							0.1	0.1					
I													
J										0.5			
K											0.5		
L												0.5	
M													0.5
N													
O													
P													
Adhesion time (minutes)	180°C	13	8	7	22	43	21	44	41	110	63	60	100
	190°C	6	3	3	10	23	9	23	21	91	44	43	87

[0016]

[Table 2]

[Continuation of Table 1]

							Comparative Examples					
No.		13	14	15	16	17	1	2	3	4	5	6
A		100	100	100	100	100	100	100	100	100	100	100
B			0.5									
C		0.3		0.5	0.5	0.5						
D		0.2										
E			0.1	0.1	0.1	0.1						
F								0.1				
G		0.1										
H				0.1					0.1			
I												
J				0.2						0.5		
K				0.3								
L					0.3							
M		0.5			0.2	0.2						
N			0.5			0.3						
O											0.5	
P												0.5
Adhesion time (minutes)	180°C	98	97	89	108	88	0	0	0	14	0	0
	190°C	86	83	78	89	78	0	0	0	7	0	0

[0017] In Table 1, A is the polyester resin Easter PETG6763 (manufactured by Eastman Chemical Co.), B is tris(octadecyl) phosphate, C is zinc bis(dioctadecyl phosphate), D is calcium dodecyl phosphate, E is butylated hydroxytoluene, F is tetrakis[methylene-3-(3,5-tert-4-hydroxyphenyl)propionate] methane, G is decyl diphenyl phosphate, H is dioctadecyl 2,2'-dimethylene trimethylene diphosphate, I is zinc dodecyl poly(oxy-propylene) phosphate, J is tris[butyl tri(oxypropylene)] phosphate, K is dodecyl octyl di-(oxyethyl) phosphate, L is bis[tetradecyl poly(oxyethylene) phosphate, M is dodecyl poly(oxyethylene) phosphate, N is zinc decyl(oxyethylene) phosphate, O is ethylene bis(stearyl phosphate), and P is zinc stearate.

[0018]

[Effect of the Invention] As described above, the polyester resin composition of this invention is obtained by means of compounding specific phosphoric acid ester compounds, for which reason calender processing of polyester resin composition, which was difficult in the past, is facilitated and products of superior quality are made possible.

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